

Remarks/Arguments

Claims 1, 69, 73, 74 and 78 have been amended. Claim 32 has been canceled. Claim 79 has been added. Claims 1-31 and 33-79 are in the application upon entry of this amendment. Entry of this amendment and reexamination and reconsideration of the present application are respectfully requested.

Claims 1, 69, 73 and 74 have been amended to include the following limitation:

the heat exchanger comprising one or more heat exchange channels and a heat exchange fluid in the heat exchange channels, the heat exchange fluid undergoing a phase change in the heat exchange channels

Similarly, claim 78 has been amended to include the following limitation:

wherein a first heat exchange fluid flows in the first set of heat exchange channels and undergoes a phase change in the first set of heat exchange channels, and another heat exchange fluid flows in the another set of heat exchange channels and undergoes a phase change in the another set of heat exchange channels, the first heat exchange fluid being the same as or different than the another heat exchange fluid.

Support for these amendments can be found in the Applicants' specification at page 4, line 26 to page 5, line 5; page 26, lines 8-13; page 29, lines 11-30; page 46, line 25 to page 47, line 11; and original claim 32.

Claims 1-59, 62-64, 68-72 and 75-78 have been rejected under 35 U.S.C. §103(a) as unpatentable over TeGrotenhuis(WO 03/078052 A1) in view of Reyes (U.S. Patent 6,726,850) and Tonkovich '536 (U.S. Patent 6,200,536). Claims 60, 61 and 65-67 have been rejected under 35 U.S.C. §103(a) as unpatentable over TeGrotenhuis, Reyes, Tonkovich '536 and Wainwright (U.S. Patent 4,366,260). Claim 73 has been rejected under 35 U.S.C. §103(a) as unpatentable over TeGrotenhuis, Reyes, Tonkovich '536 and Wentworth (U.S. Patent 4,235,799). Claim 74 has been rejected under 35 U.S.C. §103(a) as unpatentable over TeGrotenhuis in view Liu et al. ("Selective One-Step Synthesis . . .," J. Phys. Chem. B 2003, 107, 10840-10847). Claims 75-77 have been rejected under 35 U.S.C. §103(a) as unpatentable over TeGrotenhuis in view of Reyes, Tonkovich '536 and Schmidt (U.S. Patent 6,452,061). These rejections are respectfully traversed.

TeGrotenhuis discloses a microchannel device wherein a reaction chamber is in thermal contact with a heat exchange channel. The microchannel device is illustrated in Fig. 9. Referring to Fig. 9, the device includes reaction flow path 51 and heat transfer channel 61, either or both of which may include microchannels. Reactants flow through the reaction flow path from inlet 53 to outlet 55. Reaction chamber 52, which is defined by the presence of catalyst 75, can span some or substantially all of the length of the flow path 51. Heat exchange fluid flows through the heat exchange channel 61 from inlet 63 to outlet 65. Solid wall 70 separates the heat exchange channel 61 from the reaction chamber 52. (See, TeGrotenhuis at page 10, lines 3-17.)

TeGrotenhuis does not suggest the use of two or more separate reaction zones in the reaction flow path 51. In fact, the Examiner admits that "TeGrotenhuis fail to teach that an intermediate is formed in a first reaction zone with a first catalyst and a final product in a second reaction zone." In contrast, the Applicants' independent claims 1, 69, 73, 74 and 78 specify "a first reaction zone" and "another reaction zone" in the same process microchannel. Claims 1, 69 and 73 also specify the following limitation which is not suggested by TeGrotenhuis:

the first reaction zone and the another reaction zone being in the same process microchannel and being separated by a non-reactive zone in the process microchannel not containing catalyst wherein the intermediate product composition is heated or cooled.

The Applicants' amended independent claims 1, 69, 73, 74 and 78 specify the employment of heat exchange channels for exchanging heat with the reaction zones wherein a heat exchange fluid in the heat exchange channels undergoes a phase change. The employment of heat exchange fluid that undergoes a phase change enhances the heat transfer between the reaction zones and the heat exchange channels beyond that provided by convective heating or cooling. For example, for a liquid heat exchange fluid being vaporized additional heat is transferred from the reaction zones as a result of latent heat of vaporization required by the heat exchange fluid. See, Applicants' specification at page 46, lines 25-31. This is not suggested by TeGrotenhuis.

Claims 2-68 and 75-77 depend from claim 1 and are distinguishable from the teachings in TeGrotenhuis for at least the same reasons as claim 1. Claims 70-72 depend

from claim 69 and are distinguishable from the teachings in TeGrotenhuis for at least the same reasons as claim 69.

Claim 74 is directed to a process for conducting a dimethylether synthesis reaction wherein in a first reaction zone in a microchannel reactor the approach to equilibrium for the conversion of CO is from about 75% to about 95%, and the approach to the equilibrium for the conversion of CO in another reaction zone is from about 75% to about 95%. This is not suggested by the teachings in TeGrotenhuis

Claim 78 is directed to a process for conducting an equilibrium limited chemical reaction in a microchannel reactor employing a first reaction zone and another reaction zone wherein the first reaction zone is heated or cooled by a first set of heat exchange channels and the another reaction zone is heated or cooled by another set of heat exchange channels. The first set of heat exchange channels is separate from the another set of heat exchange channels. This is not suggested by the teachings in TeGrotenhuis

In the rejection of claims 1-59, 62-64, 68-72 and 75-78, the Examiner contends that TeGrotenhuis "teach a process for oxidation (page 27) wherein the reactions are equilibrated to 90% conversion (page 13) and wherein sequential reactors, having ribs formed of catalysts (fig. 10, #164), are used in the process (page 26) wherein the multiple microchannels have temperatures around 300°C (page 31)." Similarly, in the rejection of claim 73 the Examiner contends that TeGrotenhuis "teach a process for methanol synthesis (page 27) wherein the reactions are equilibrated to 90% conversion (page 13) and wherein sequential reactors are used in the process (page 26) wherein the multiple microchannels have temperatures around 300°C (page 31)." Applicants respectfully disagree with these contentions.

The process disclosed on page 13 where a 90% conversion is referred to is a water gas shift (WGS) reaction, not an oxidation reaction or a methanol synthesis reaction as indicated by the Examiner. Also, the process disclosed on page 31 where a 300°C temperature is referred to is a WGS reaction.

The 90% conversion referred to on page 13 was taken from the plot shown in Figure 1 which is for a WGS reaction. This plot indicates that the reaction rate for the WGS reaction dropped "by over three orders of magnitude by the time 90% conversion is reached." See, TeGrotenhuis at page 13, lines 17-22. The 90% conversion value referred

to in this passage is not the same as the "approach to equilibrium" which is specified in the Applicants' independent claims 1, 69, 73, 74 and 78. The term "approach to equilibrium" is defined in the Applicants' specification at page 20, line 35 to page 21, line 9, as being the actual conversion of a reactant divided by the equilibrium conversion for that reactant.

The Examiner's contention that TeGrotenhuis discloses "sequential reactors, having ribs formed of catalyst (Fig. 10, #164)" is not correct. Applicants respectfully submit that the cited passage from TeGrotenhuis discloses a method for making a microchannel reactor, but does not suggest the use of sequential reactors as contended by the Examiner. In Figs. 8 and 10-12, TeGrotenhuis discloses microchannel reactor 100 which includes reactant inlet 110, reactant outlet 120, heat exchange fluid inlet 130 and heat exchange fluid outlet 140. Reactor 100 is constructed by alternatively stacking a series of shim sheets designated A (Fig. 10), B (Fig. 11) and C (Fig. 12). The shim sheets A, B and C contain channel features and when assembled provide a stacked array of microchannel flow paths in reactor 100. Each of the shim sheets A, B and C contains three identical shim patterns so that three reactors 100 can be constructed from the shim sheets. See, TeGrotenhuis at page 20, line 8 to page 21, line 4. The cited passage does not suggest the use of sequential reactors as contended by the Examiner.

The Examiner cites Reyes against claims 1-73 and 75-78 for its disclosure of a multistage process. Reyes discloses a multistage catalytic partial oxidation process for oxidizing a hydrocarbon feedstream comprising C₁-C₄ hydrocarbons, with an oxygen-containing feedstream to produce a product comprising CO and H₂, also known as synthesis gas. The total oxygen requirement for the process is introduced incrementally, the first of the incremental additions taking place in the first reaction stage, and subsequent incremental additions taking place in each of the subsequent reaction stages. Each reaction stage contains a partial oxidation catalyst. The reference does not suggest conducting the disclosed process in a microchannel, as specified in the Applicants' claims 1-33, 35-73 and 75-78. In fact, the examples disclosed in the reference specify the use of a quartz reactor containing a foam support for the catalyst, the dimensions of the foam support being 0.75 inch (19.05 mm) in diameter by 0.5 inch (12.7 mm) in length. See, Reyes at column 12, lines 52-53; column 13, lines 45-46; and column 14, lines 44-45. The foam supports disclosed in these examples are clearly too large for the microchannel

reaction zones specified in the Applicants' claims 1-33, 35-73 and 75-78. The reference contains no disclosure relating to determining the equilibrium conversion value for a reactant and conducting the reaction in a microchannel wherein each reaction stage is in the same microchannel and the approach to equilibrium is at least about 5% as specified in the Applicants' independent claims 1 and 78, or at least about 40% as specified in the Applicants' independent claim 69. Independent claim 73 is distinguishable from the teachings in this reference by specifying a process for making methanol, which is not suggested by the teachings in Reyes.

The Examiner admits that "TeGrotenhuis fails to teach that the reaction zones are separated by a non-reactive zone, the first reaction zone and another reaction zone being in the same process microchannel." The Examiner attempts to make up for this deficiency by relying on the disclosure in Fig. 2(d) of Tonkovich '536. The Examiner contends that Fig. 2(d) in Tonkovich '536 discloses "a microchannel reactor configuration . . . wherein a heat exchanger is disposed between two reactors within a single process microchannel." This is not an accurate interpretation of the disclosure shown in Fig. 2(d). In Fig. 2(d), Tonkovich '536 shows a reactor with an exothermic reaction chamber 100 and a second reaction chamber 230. These reaction chambers are separated by exhaust chamber 108, heat exchanger 114 and heat transfer channel 220. These reaction chambers are connected by exhaust outlet 112. Fig. 2(d) of Tonkovich '536 does not show a microchannel reactor containing two reaction zones in the same microchannel as specified in the Applicants' claims 1-31, 33-73 and 75-78.

Wainwright et al. is cited against claims 60, 61, 65-67 to support the contention that it is known that specific catalysts are capable of producing both methanol and dimethyl ether depending on the conditions of the reactions. Wainwright et al. discloses a catalyst which is used to produce methanol or a mixture of methanol and dimethyl ether. However, Wainwright contains no disclosure that would suggest the limitations of the Applicants' claims 60, 61 and 65-67. The Applicants' claim 60 specifies that the contact time of the reactant composition and/or intermediate product composition with the catalyst in the first reaction zone is from about 10 to about 500 milliseconds. Claim 61 specifies that the contact time of the intermediate product composition and/or product with the catalyst in the

another reaction zone is from about 10 to about 500 milliseconds. Neither of these are suggested by Wainwright. Claim 65 specifies that the pressure within the process microchannels is at least about 1 atmosphere, which is not suggested by Wainwright. Claim 66 specifies that the pressure drop for the flow of the reactant composition and product through the process microchannels is up to about 40 atmospheres per meter of length of the process microchannels. This is not suggested by Wainwright. Claim 67 specifies that the pressure drop for heat exchange fluid flowing through the heat exchange channel is up to about 50 atmospheres per meter of length of the heat exchange channel. This is not suggested by Wainwright.

Wentworth is cited against claim 73 for its disclosure of "a method for making methanol wherein a reactant composition is passed through multiple catalyst reactions for the purpose of minimizing the amount of heat exchange required to heat the inlet gas to a temperature appropriate for the synthesis reaction to initiate." The process disclosed in Wentworth involves the use of a plurality of catalysts arranged in sequence. The gasses pass over the catalyst at successively higher temperatures, then are cooled, and then passed into one or more catalyst beds at lower temperatures (column 1, lines 58-61). Wentworth does not, however, suggest conducting the process in a microchannel as specified in claim 73. In fact, the catalyst volumes disclosed in Wentworth range from 500 cubic feet to 1250 cubic feet (column 10, lines 12-16) which are clearly not suitable for use in a microchannel as specified in claim 73. Similar catalyst volumes are disclosed in columns 12 and 15 (600-1600 cubic feet), column 17 (400-1500 cubic feet), column 19 (2000-3000 cubic feet), and column 21 (300-1200 cubic feet). Also, the reference indicates that the catalyst "would ordinarily be one-fourth inch (0.635 mm) by one-fourth inch (0.635 mm) right cylinders" (column 10, lines 23-25). These dimensions are not suggestive of catalysts suitable for use in a microchannel as specified in claim 73.

In citing Liu et al. against claim 74, the Examiner contends that Liu et al. discloses the production of dimethylether in a microreactor. This is not correct. The disclosure in Liu et al. relates to the production of dimethoxymethane by the oxidation of dimethylether or methanol using as the catalyst unsupported or silica-supported heterolpolyacids with Keggin structures. Thus, the reference is directed to the use of dimethylether as a starting

material, not the production of dimethylether. The reference does not suggest the production of dimethylether in a microchannel as specified in the Applicants' claim 74.

The Examiner admits that TeGrotenhuis fails to teach a method for making dimethyl ether, but cites Shikada against claim 74 contending that it "teach a method of making dimethylether (col. 1) wherein carbon monoxide and hydrogen are flowed through a two reactor process wherein dimethylether is produced in both reactors (col. 4)." Shikada discloses an apparatus for producing dimethylether comprising: a slurry-bed reactor filled with a dimethylether synthesis catalyst and a medium oil therefor; a condenser for condensing a gasified medium oil discharged from the reactor; an adsorber for removing a catalyst-deactivation ingredient from the medium oil condensed in the condenser; and recycle means for recycling the medium oil to the slurry-bed reactor. This is not the same as the process specified in the Applicants' claim 74 wherein the reaction is conducted in a microchannel, a first reaction being conducted in a first reaction zone in the microchannel and another reaction being conducted in another reaction zone in the same microchannel.

In the rejection of claims 75-77, the Examiner contends that if TeGrotenhuis fails to teach the limitations of these claims, such limitations are taught by Schmidt. In this rejection the Examiner contends that Schmidt et al. teach a method of oxidation of hydrocarbons wherein the claimed SLPM and contact time is known for conversion reactions. Schmidt et al., however, does not suggest conducting the reaction in a microchannel reactor comprising at least one process microchannel wherein a first reaction zone and another reaction zone are positioned in the same process microchannel and these reaction zones are separated by a non-reactive zone not containing catalyst as specified in claims 75-77. In fact, the reactor used in the examples disclosed in Schmidt was a quartz tube with an inside diameter of 18 mm (see, column 5, lines 10-12), which is clearly not a microchannel as specified in the Applicants' claims 75-77.

Withdrawal of the rejection of claims 1-33 and 35-78 is believed to be warranted and is respectfully requested.

Applicants respectfully submit that the application is in condition for allowance. A Notice of Allowance is respectfully requested. In the event the Examiner would like to discuss any matters concerning this application, he is invited to contact the undersigned

attorney by telephone. Any fees required for the filing of this paper may be charged to Deposit Account Number 18-0988.

Respectfully submitted,

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